Effects of Spectral Dispersion of Cloud Droplet Size Distributions on Radiative Properties of Clouds and Dispersion Forcing

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Introduction

Most studies of the indirect aerosol effect on cloud radiative properties have considered only changes in N caused by increasing the cloud condensation nucleus (CCN) concentration. In such studies, it is assumed that the change in r_e, due to the increase in N, can be calculated from the simple relation

$$r_e = [3/(4\pi\rho_w)]^{1/3} (L/N)^{1/3}$$
 (1)

where L is the liquid water content, and ρ_w is the density of water. In effect, these studies implicitly assume the only effect of aerosols on cloud microphysical properties is to increase N, and r_e can be parameterized by Eq. (1) for monodisperse droplet size distributions. Eq. (1) has also been used in the parameterization of r_e . However, available data suggests the spectral shape of the cloud droplet size distribution may also be influenced by the addition of anthropogenic aerosols, and our recent studies, together with others, have shown changes in d have a significant effect on calculation of r_e (Pontikis and Hicks 1992; Martin et al. 1994; Liu and Hallett 1997; Liu and Daum 2000). This paper expands on these previous studies to address the effect of d on cloud radiative properties, and the role anthropogenic aerosols may have in determining d of marine clouds.

Spectral Dispersion and Its Effect on Cloud Effective Radius, Cloud Optical Depth, and Cloud Albedo

The value of r_e can be generally parameterized as

$$r_e = \alpha [L/N]^{1/3} \tag{2}$$

where α is a parameter that has been shown to depend on d (Pontikis and Hicks 1992; Martin et al. 1994; Liu and Hallett 1997; Liu and Daum 2000). For purposes of examining the effect of d on r_e and subsequently on cloud radiative properties we use as a reference, a cloud with a monodisperse size

distribution where r_e is parameterized by Eq. (1). Accordingly we define the dimensionless quantity β as

$$\beta = \alpha/\alpha_0 \tag{3}$$

where α is the prefactor corresponding to a given d, and α_0 is the prefactor for the monodisperse reference cloud with the same N and L $[\alpha_0 = (3/4\pi\rho_w)^{1/3}]$. A review of the various ways of specifying the relationship between β and d, is given in Liu and Daum (2000) and Daum and Liu (2000). As shown in Figure 1, the Weibull (or Gamma) droplet size distribution yields the most accurate β -d relationship for both continental and marine stratiform clouds. Subsequent calculations of the dependence on d of r_e and cloud radiative properties will be made assuming the dependency between d and β follows that exhibited in Figure 1.

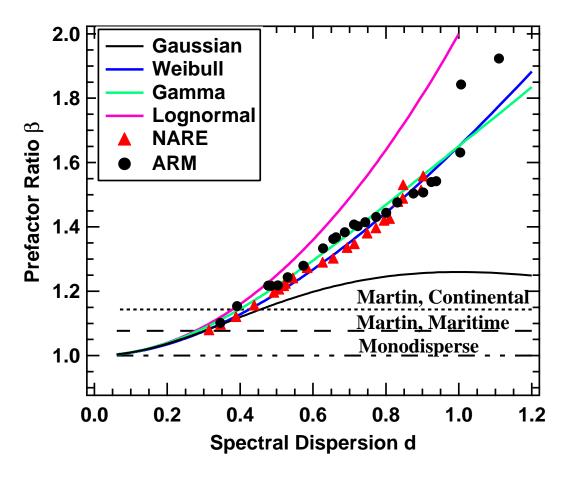


Figure 1. The plot of β versus d shows that the Weibull (Gamma) droplet size distribution best represents the relationship between b and d exhibited by the ambient data. The lognormal distribution tends to overestimate, and the Gaussion distribution tends to underestimate b. The data labeled NARE are from marine stratiform clouds measured during the 1993 North Atlantic Regional Experiment; the data labeled Atmospheric Radiation Measurement (ARM) Program are from continental stratocumulus clouds measured during over the ARM Southern Great Plains (SGP) site during two 1997 intensive operational periods (IOPs).

Using Eqs. (2) and (3), the relative changes due to d in r_e , τ , and R are respectively,

$$E_{r} = \frac{r_{e} - r_{e0}}{r_{e0}} = \beta - 1 \tag{4}$$

$$E_{\tau} = \frac{\tau - \tau_0}{\tau_0} = \frac{1 - \beta}{\beta} \tag{5}$$

$$E_{R} = \frac{R - R_{0}}{R_{0}} = \frac{(1 - \beta)(1 - R_{0})}{R_{0} + (1 - R_{0})\beta}$$
 (6)

where the subscript "0" denotes values for the reference cloud. In deriving the above equations, the following expressions are used:

$$\tau = \frac{3H}{2\rho_{\rm w}} \frac{L}{r_{\rm e}},\tag{7}$$

and
$$R = \frac{(1-g)\tau}{2+(1-g)\tau}$$
, (8)

where H is the cloud thickness and g is the asymmetry factor.

Figure 2 shows E_r and E_τ as a function of d. It is clear from this figure that for a cloud with a given N and L, an increase in d causes an increase in r_e and a corresponding decrease in τ . The changes in both r_e and τ are small at low values of d, but increases non-linearly as d increases. Figure 3 shows E_R as a function of d for different values of R_0 . As indicated by this figure, the effect of d on E_R is both a function of R_0 and d. When the cloud is totally reflective, i.e., $R = R_0 = 1$, and d has no effect. For values of $R_0 < 1$, the relative difference in R increases with increasing d. The effect gets larger as R_0 decreases, and for a cloud with an R_0 between 0.3 and 0.7 (typical for ambient clouds); the relative difference may be as large as 20 percent. Differences of this magnitude are significant in calculation of the energy balance of a cloudy sky.

Spectral Dispersion and Cloud Droplet Number Concentration

To support the notion that increasing aerosol concentrations in the marine environment may cause increases in d, in addition to increasing N and reducing the droplet radius, we show in Figure 4, a plot of d as a function of N for marine clouds. The data are taken from several studies (see the figure caption). Although the d-N relationship shown in Figure 4 is noisy, clearly there is a substantial increase in d as N increases and in most cases there is a clear difference between the group of clouds classified as clean/marine (blue symbols) and those classified as polluted/continental (red symbols) by the authors of those publications. We note the d-N relationship is expected to be noisy because there are a number of factors (e.g., turbulence) that influence d other than aerosol loading. The data shown in Figure 4 may be

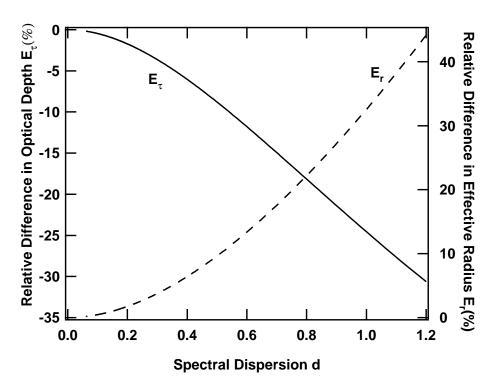


Figure 2. Relative differences (with respect to the reference cloud with a monodisperse cloud droplet size distribution) in effective radius and cloud optical depth as a function of the spectral dispersion.

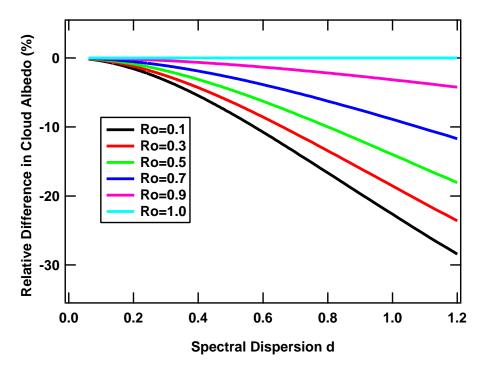


Figure 3. Relative difference (as above) in cloud albedo as a function of spectral dispersion for different values of R_0 .

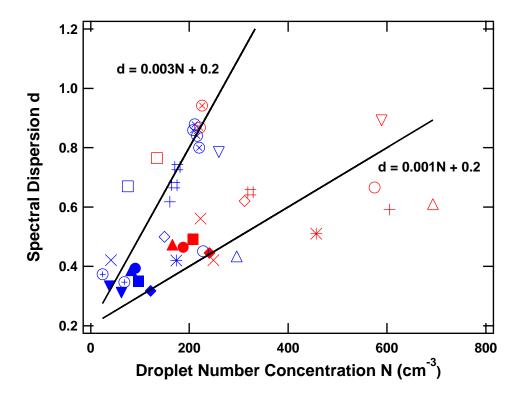


Figure 4. Relationship between spectral dispersion (d) and droplet concentration (N). Blue symbols indicate clouds classified as clean by the authors and red symbols indicate clouds as polluted or continentally influenced. • = ASTEX, Yum and Hudson (2000); ▼ = FIRE, ACE1 Yum and Hudson (2000); ◆ = SCMS, Yum and Hudson (2000); ♠ = SOCEX, Yum and Hudson (2000); ♠ = Sounding, Hudson and Yum (1997); ■ = Horizontal, Hudson and Yum (1997); ∇ = Level 1, Noonkester (1984); \triangle = Level 2, Noonkester (1984); \triangle = Level 3, Noonkester (1984); \triangle = Hudson and Yum (2000); \mathbb{R} = Vertical, Garrett and Hobbs (1995); \mathbb{R} = Horizontal, Garrett and Hobbs (1995); \mathbb{R} = Astex, Hudson and Li (1995); \mathbb{R} = Ackerman et al. (2000); \mathbb{R} = (Noone et al. 2000a); \mathbb{R} = Noone et al. (2000b).

used as support for the Twomey effect as clouds classified as polluted exhibit much higher N and smaller mean radii compared to those classified as clean. Other factors being constant, this increase in N should lead to an enhancement in R. However, the increase in N is also accompanied by an increase in d. For a given L and N, this will cause an increase in r_e and a decrease in R. In effect, the increase in d with N tends to diminish the R increase due to the increase in N. If the change in d is large enough, the enhancement in R caused by the Twomey effect will be significantly reduced and perhaps entirely cancelled by the effect of d. In the next section, we will explore the relative importance of these two effects.

Spectral Dispersion and Cloud Radiative Forcing

The global-mean radiative forcing that would result from a perturbation in R caused by a change of N has been assessed by Charlson et al. (1992) and many others using similar arguments (Kaufman et al. 1991; Schwartz 1988; Schwartz and Slingo 1996). The equation for the simple number forcing alone can be generalized to include the effects of both N and d, viz,

$$\Delta F = -\frac{0.8}{4} A_{mst} F_T \frac{R(1-R)}{3N} \Delta N + \frac{0.8}{4} A_{mst} F_T \frac{R(1-R)}{1} \frac{\partial (\ln \beta)}{\partial d} \Delta d$$
 (9)

where A_{mst} is the fraction of the globe covered by marine stratiform clouds and F_T is the solar constant. The first term on the right side of Eq. (9) expresses the traditional number forcing associated with the increase in R due to the increase in N. The second term expresses the effect due to changes in d, and represents what we define here as the dispersion forcing term. If d does not change with aerosol loading, the second term in Eq. (9) goes to zero, and Eq. (9) reduces to the traditional expression for number forcing. Note that the dispersion forcing term has a positive sign indicating that it acts to decrease R, whereas the number forcing term acts to increase R. Rearranging Eq. (9) to express (ΔF) as a function of the relative perturbation of N ($\Delta N/N$), we derive

$$\Delta F == \frac{0.8}{4} F_{\rm T} A_{\rm mst} R (1 - R) \frac{\Delta N}{3N} (-1 + r)$$
 (10)

where r is the magnitude ratio of the dispersion forcing to the number forcing and is related to the d-N relationship by

$$r = 3N \frac{\partial (\ln \beta)}{\partial d} \frac{\Delta d}{\Delta N} \tag{11}$$

The dependence of d on N is needed to calculate the global-mean shortwave forcing including both number and dispersion effects. For this purpose, we bound the dependency by deriving two linear d-N relationships which approximately represent the lower and upper limits of the data given in Figure 4, corresponding to d = 0.001N + 0.2, and d = 0.003N + 0.2, respectively. Figure 5 shows the number forcing, dispersion forcing, and total forcing as a function of N, assuming the two different relationships between d and N adduced above, ΔN/N, A_{mst}, R and F_T, were set to the same values as used in Charlson, et al. to estimate the number forcing, i.e., $A_{mst} = 0.3$, R = 0.5, and $F_T = 1370 \text{Wm}^{-2}$. As discussed in Charlson et al., the fixed fractional perturbation in number concentration (15 percent) results in about -1 wm⁻² for the number forcing term independent of N. In contrast, dispersion forcing is always positive, and increases at a rate determined by both N and the dependence of d on N. Since total cloud forcing due to enhancement CCN concentrations is the sum of the dispersion and number forcing terms, the inclusion of the dispersion forcing alters the simple "static" picture of the number forcing, and the total forcing depends on both N and the dependence of d on N. If we assume the weakest dependence of d upon N, the transition between cooling and warming occurs at N ~750/cm³; for the strongest dependence, the transition occurs at an N of about 200 cm⁻³. At all values of N the effect of dispersion forcing is to reduce the cooling predicted by the Twomey effect.

Summary and Conclusions

The arguments and data presented above demonstrate the importance of d and its relationship with N in quantifying various cloud radiative properties and cloud radiative forcing. The conventional assumption that the shape of the cloud droplet size distribution (e.g., d) does not change as a function of N can lead

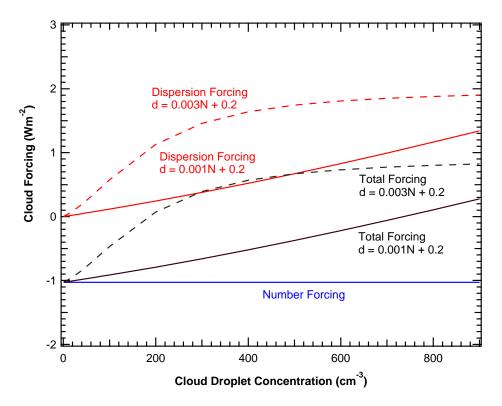


Figure 5. Number forcing (blue), dispersion forcing (red) and total forcing (black) as a function of droplet concentration for the two linear d-N relationships. The dashed and solid lines represent d = 0.003N + 0.02, and d = 0.001N + 0.02, respectively.

to significant biases in calculation of r_e , τ , R, and cloud radiative forcing. The magnitude of this effect is, however, highly uncertain, and requires additional data and analysis of the d-N relationship. If, after further study, a strong consistent relationship between d and N is found, there could be a significant change our assessment of the importance of the indirect effect of aerosols on climate.

Acknowledgement

This work is supported by the U.S. Department of Energy, Grant DE-AC02-98CH10886. We are indebted to Dr. S. E. Schwartz at Brookhaven National Laboratory for his discussions and the suggestion of using the name "dispersion forcing."

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